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Volcanic fronts as a consequence of serpentinite dehydration in the fore-arc mantle wedge

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Abstract

Serpentinities exhumed from mantle wedges are enriched in elements that are mobile at low temperatures, such as As, Sb, Pb, and Sr, based on samples from the Himalayas and the Alps. Such data provide compelling evidence that these fluid-soluble elements were incorporated into the overlying fore-arc mantle wedge from the subducting slab. Thus, serpentinites in the fore-arc mantle act as a sink for water and fluid-soluble elements, insulating the overlying mantle from further hydration.. Downward movement of the serpentinite layer, facilitated by mantle flow, causes transport of these elements to deeper, hotter levels in the mantle. Eventual dehydration of these serpentinites discharges the fluid-soluble elements with water, leading to partial melting of the mantle wedge and accounting for the observed enrichment of these elements in magmas at the volcanic front.

keywords: subduction zones, volcanic arcs, chalcophile elements, serpentinite, eclogite, fore-arc mantle, geochemical cycles.

Introduction

Active arc volcanoes are typically arranged parallel to the trench, where the subducting slab has reached a depth of ~80 to 140 km. The intervening fore-arc region, more than 100 km wide, is essentially barren of volcanoes (e.g., Gill, 1981; Tatsumi, 1986). Volcanism at the volcanic front is generally attributed to the rapid release of water from subducting slabs as they are metamorphosed to eclogite facies (Tatsumi, 1986; Peacock,

1993; 1999). Water released from subducting slabs migrates upwards, forming amphiboles in the overlying mantle, and the subsequent break-down of the amphiboles leads to the partial melting that drives arc volcanism (Gill, 1981; Tatsumi, 1986, 1989).

Slabs and overlying sediments continuously release water during subduction (e.g., Schmidt and Poli, 1998; Mibe et al., 1999). Why then does this not lead to partial melting and volcanism in the fore-arc, given that the temperature of the mantle in this region, over 1200 °C (e.g., Furukawa, 1993), is sufficient for partial melting? In addition, the formation of eclogite, causing water release, occurs at a shallower depth than that corresponding to the position of the volcanic front (Fukao, 1983). Furthermore, volcanic-front magmas are enriched in As, Sb, Pb, and B (Hofmann, 1988; Ryan et al., 1995; Noll et al., 1996), elements that are mobile even at surface temperatures (Thomson et al., 1993; Smeldley and Kinniburgh, 2002). How can these mobile elements be retained in descending slabs until they are transformed to eclogite? This paper presents the evidence for the enrichment of fluid-soluble elements in serpentinites in the fore-arc mantle, an elemental assemblage similar to that of volcanic-front magmas. We propose that partial melting for these magmas is related to the hydration and subsequent dehydration of peridotites in mantle wedges, instead of direct dehydration of subducting slabs.

Samples

The serpentinite samples for this study are all associated with eclogites, and were exhumed from mantle wedges together with the eclogites (Guillot et al., 2000; 2001). Samples were collected from the Ladakh area of the northwest Himalayas, associated with the eclogitic Tso Moriri unit. The Tso Moriri unit represents the subducted margin of the Indian continent, which reached a depth of ~ 100 km and a temperature of ~ 600°C before it was exhumed, together with the serpentinites, in the Early Miocene (Guillot et al., 2000; 2001). Thus, the serpentinites represent hydrated mantle peridotites overlying the subduction zone.

Samples from the Alps and Cuba are similar in their geological occurrence. They are associated with omphacitic eclogites, a metamorphic product of subducted oceanic crust. The

Alps samples were collected near Mont Viso in northwestern Italy, and the Cuban samples from Zaza in central Cuba, part of the Caribbean eclogites (Mann, 1996). The serpentinite samples are similar in refractory major-element composition to the Himalayan samples. They also formed by hydration of the mantle wedge, and were exhumed together with the eclogites (Blake et al., 1995). Voluminous oceanic peridotites in these areas (e.g., Philippot, 1993) were not used in the study.

Chemical composition of serpentinites

Serpentinites consist mainly of antigorite and minor chrysotile, chlorite, talc, and Mg-rich olivine, as well as primary chromite. All of our serpentinite samples have a similar bulk chemical composition, with high concentrations of compatible elements, such as Mg and Cr; they also show a gradual decrease in concentrations from highly compatible Mg to moderately compatible Zr (in order starting from the right side of Fig. 1). Such pattern confirms our earlier interpretation, based on chromite chemistry and the content of platinum group elements, that they were derived from a refractory mantle wedge (Guillot et al., 2000; 2001).

A refractory mantle should contain low concentrations of Pb, As, Sb and light rare-earth elements (REE) because they are preferentially removed from the mantle during partial melting. However, our serpentinite samples, particularly the Himalayan samples, show an extreme enrichment in As (up to 130 ppm), Sb (up to 12 ppm), Pb (up to 23 ppm), Sr (up to 56 ppm) and light REE (up to 0.54 ppm Ce) (Fig. 1). The actual degree of enrichment was much greater than that shown in the diagram because their concentrations in a refractory mantle are much lower than the primitive mantle values that are used for normalization (Fig. 1). For example, the abundance of As and Sb in the refractory mantle is deduced to be 14 ppb and 1.7 ppb, respectively (Noll et al., 1996), which is one third the amount in the primitive mantle (McDonough and Sun, 1995).

Cause for the enrichment of selected elements

A common character among the enriched elements is their high solubility in aqueous fluids (e.g., Rose et al., 1979; Smeldley and Kinniburgh, 2002), suggesting that the

serpentinites incorporated these elements during hydration of mantle peridotites. It is not surprising that the Himalayan samples show a high enrichment of these elements (Fig. 2), as the subducted slab is the northern margin of Indian continent is comprised of Archean granitoids and clastic sediments derived from the granitoids. Granitic rocks contain higher concentrations of Sb, As, Pb and Sr than oceanic crust (Rose et al., 1979). Such incorporation is also supported by the isotopic signatures of these serpentinites. The values of $^{143}\text{Nd}/^{144}\text{Nd}$, expressed as ϵNd , are as low as -20 (18), and $^{87}\text{Sr}/^{86}\text{Sr}$ values are as high as ~ 0.720 (our unpublished data), similar to Archean granitic rocks. This indicates that Sr and Nd were released from the subducting sediments on the margin of the Indian continent and incorporated in the overlying mantle peridotite during serpentinization (Guillot et al., 2001).

Physical mixing of crustal rocks with serpentinites may have caused such an enrichment, as tectonic melanges are common along subduction zones. However, this possibility is rejected because the compositional pattern of serpentinites is very different from that of continental crust (Fig. 1).

Arsenic, Sb, and Pb are chalcophile, suggesting that sulfides may have some role in the enrichment. All serpentinite samples, however, contain less than 0.01 wt % S, and other chalcophile elements, such as Cd are not enriched. In addition, Sr and Ce, elements unrelated to sulfides, are also enriched. Therefore, S did not contribute to the observed enrichment.

Timing and temperature of enrichment

The elements enriched in serpentinites are all mobile in low-temperature aqueous fluids, based on their common enrichment in groundwaters and organic-rich sediments (e.g., Thomson et al., 1993), and low-temperature (100 °C) thermal springs (Weissberg et al., 1979). Arsenic and Sb are easily reduced from As^{5+} and Sb^{5+} to soluble As^{3+} and Sb^{3+} during compaction and burial of sediments, and lead is easily released from sediments during diagenesis at temperatures below 150 °C (Hanor, 1979). Mississippi valley-type Pb-Zn deposits, a major source of Pb, formed at less than 150 °C (e.g., Hanor, 1979, Sverjensky, 1986). These data indicate that As, Sb and Pb could be mobilized and discharged from subducting slabs at low temperatures during the early stage of subduction.

Such mobility is supported by the composition of subducted sediments which now form the Catalina schist. The contents of As and Sb sharply decrease in epidote-blueschist and epidote-amphibolite facies rocks compared to lawsonite-albite facies rocks (Bebout et al., 1999), suggesting that they were released during early metamorphism at temperatures below 300 °C.

Tin and Mo have a compatibility similar to light REE, As, and Sb (Jochum et al., 1993). However, Sn and Mo are not mobile in low-temperature fluids (Seby et al., 2001), as demonstrated by the high-temperature, > 500 °C, formation of most Sn and Mo deposits in igneous rocks (e.g., Roedder, 1984). Therefore, the enrichment of As, Sb, and Pb without significant enrichment of Sn and Mo in our serpentinite samples suggests that the incorporation of As, Sb, and Pb, and the associated hydration of serpentinites likely took place at low to moderate temperatures, < 400 °C.

Recent geophysical data suggest the occurrence of a serpentinite layer at the base of the mantle wedge in the fore arc (Furukawa, 1993; Kamiya and Kobayashi, 2000). Direct evidence for the presence of serpentinites is provided by serpentinite diapirs in fore-arc regions, such as the Mariana arc (Fryer, 1996). A recent boron isotope study of these serpentinites confirms that the mantle peridotites hydrated at relatively low temperatures, < 300 °C (Benton et al., 2001).

Composition of arc magmas

Arc magmas are characterized by high concentrations of large-ion-lithophile elements (LILE) (e.g., Gill, 1981). The enriched elements comprise not only lithophile elements, but also fluid-soluble chalcophile elements such as Pb, As and Sb (Hofmann, 1988; Ryan et al., 1995; Noll et al., 1996). The ratios of As/Ce, Sb/Ce and Pb/Ce of arc magmas are much higher than those of mid-oceanic ridge basalts (MORB) and oceanic island basalts (OIB) (Fig. 2), illustrating the enrichment of As, Sb and Pb in the mantle source region of arc magmas. The extent of enrichment at the source is evaluated by comparing arc magmas with MORB (Fig. 1), as the comparison of magmas eliminates the effects of melt formation. This shows that the source is enriched in the following order: As, Sb, Pb and Ce. The enrichment

of the source mantle is similar to that of the serpentinites (Fig. 1), suggesting a linkage between serpentinite composition and generation of arc magmas.

Proposed model

Altered and hydrated oceanic basalts and sediments contain high concentrations of As, Sb, and Pb (Plank and Ludden, 1992; Jochum and Verma, 1996). Water and these soluble elements are expelled from slabs during the early stages of subduction. These components migrate upward and hydrate the mantle peridotite to form a serpentinite layer along and just above the subduction zone (Fig. 3). The dissolved elements become highly concentrated as the water is consumed by hydration, and are eventually fixed in the serpentinites when the water is totally consumed.

Serpentinites contain over 13 wt % H₂O in mineral structures, providing a sink for the flux of water during subduction dehydration. Volume increase during peridotite hydration also prevents water from permeating into the interior of the mantle wedge. Therefore, serpentinite layers act as a barrier, keeping the interior of the mantle wedge dry and free from partial melting, even though it is sufficiently hot to partially melt if hydrated (e.g., Furukawa, 1993).

The serpentinite layers are dragged downward by mantle flow (Furukawa, 1993), resulting in trace elements that are mobile at low temperatures being transferred to the deep mantle wedge. When the temperature reaches ~ 650 °C, serpentinites decompose to forsterite and enstatite (Ulmer and Trommsdorff, 1995; Wunder et al., 2001). This corresponds to ~ 90 to 110 km depth in many arcs, such as Cascadia, southwestern Japan, and southern Chile (Fig. 4).

By contrast, old subducting slabs are cold and the subduction planes may not reach 650 °C, even at 130 km depth. However, such subduction zones contain thick serpentinite layers above the subduction planes because of an extended period of water supply. The thickness of the serpentinite layer in northeast Japan, the longest-lasting subduction zone in the world, is estimated to be greater than 10 km in the fore-arc region (Furukawa, 1993). The bottom of the serpentinites in contact with a cold slab is cold, but the top is much hotter.

Serpentinites ~ 10 km above the subduction plane in northeastern Japan at 130 km depth start dehydrating at 600-650 °C (Peacock, 2001).

The break-down of serpentinites releases a large amount of water as well as elements that are not compatible with olivine. The water and fluid-soluble elements migrate upwards, and are incorporated by the partial melt that generates arc magmas.

The widely held view of the formation of arc magmas involves a rapid release of water from subducting slabs during the transition from amphibolite/blueschist to eclogite (e.g., Tatsumi, 1986, 1989; Peacock, 1993; Peacock and Wang, 1999). The water forms amphiboles in the overlying mantle wedge, and their break-down leads to partial melting. Several workers have noted the problems associated with this model, including the shallow depth for eclogite formation (Fukao et al., 1983; Schmidt and Poli, 1998; Mibe et al., 1999). The discrepancy in depth between that of eclogite formation and frontal-arc magma generation is over 30 km in northeastern Japan (Fukao et al., 1983). Such discrepancy may be attributed to downward flow of amphibole peridotite, but this implies a delay of dehydration by more than 600,000 years, using a steep subduction angle of 30 ° and a fast subduction rate of 10 km/year. This seems to be an unreasonably long time for the dehydration of amphiboles.

Other explanations for the lack of magmatism in forearc regions include mechanical difficulties involved with the ascent of magma and aqueous fluids (Mibe et al., 1999; Schmidt and Poli, 1998). Experiments indicate no upward movement of aqueous fluids at low pressures, < 2 GPa, in the mantle (Schmidt and Poli, 1998). However, the static experimental conditions are quite different from the natural environment, where fluid flow is commonly driven by gradients in fluid pressure and under conditions of high stress. Abundant evidence indicates that fluids move along fault and shear zones at deep crustal to mantle depths (e.g., Cathles, 1990). In addition, the occurrence of serpentinites in the fore-arc mantle (Fryer, 1996) attests to the movement of aqueous fluids under relatively low pressures in the mantle.

Our model is consistent with the across-arc geochemical variations of magmas. The enrichment of As, Sb and Pb is strongest at the volcanic front and decreases sharply with increasing distance from the volcanic front (Ryan et al., 1995; Noll et al., 1996). Serpentinites

incorporate these elements at low temperatures, but retain them only as long as serpentinites are stable. Once the serpentinites are destabilized, they are discharged with aqueous fluids, dissipated upwards into the mantle and partially incorporated in melt.

In addition, our model also accounts for the different behavior observed among high field-strength elements (HFSE; elements with large valence and small ionic radii) in subduction zones. Low concentrations of HFSE are considered to be the key characteristic feature of arc magmas (Gill, 1981) and are generally thought to be incorporated into rutile in subducting slabs, and thus are not available for arc magmas. Antimony is a HFSE with similar ionic radius to Nb and thus should be low in arc magmas, but the two are decoupled (the so-called “Sb paradox”; Zack et al., 2002). The concentration of Sb is high in arc magmas, particularly in frontal arc magmas (Noll et al., 1996). Our model would have soluble Sb released from the subducting slabs at low temperatures, prior to being fixed by rutile formation in slabs (Fig. 3). The soluble Sb is transferred to the overlying peridotite, leading to its eventual enrichment in frontal arc magmas. On the other hand, Nb and Ta are insoluble at low temperatures and remain in the slabs, to be incorporated into metamorphic rutile. Thus, the different behaviour of HFSE in low-temperature fluids can explain their decoupling and disparate fates during subduction.

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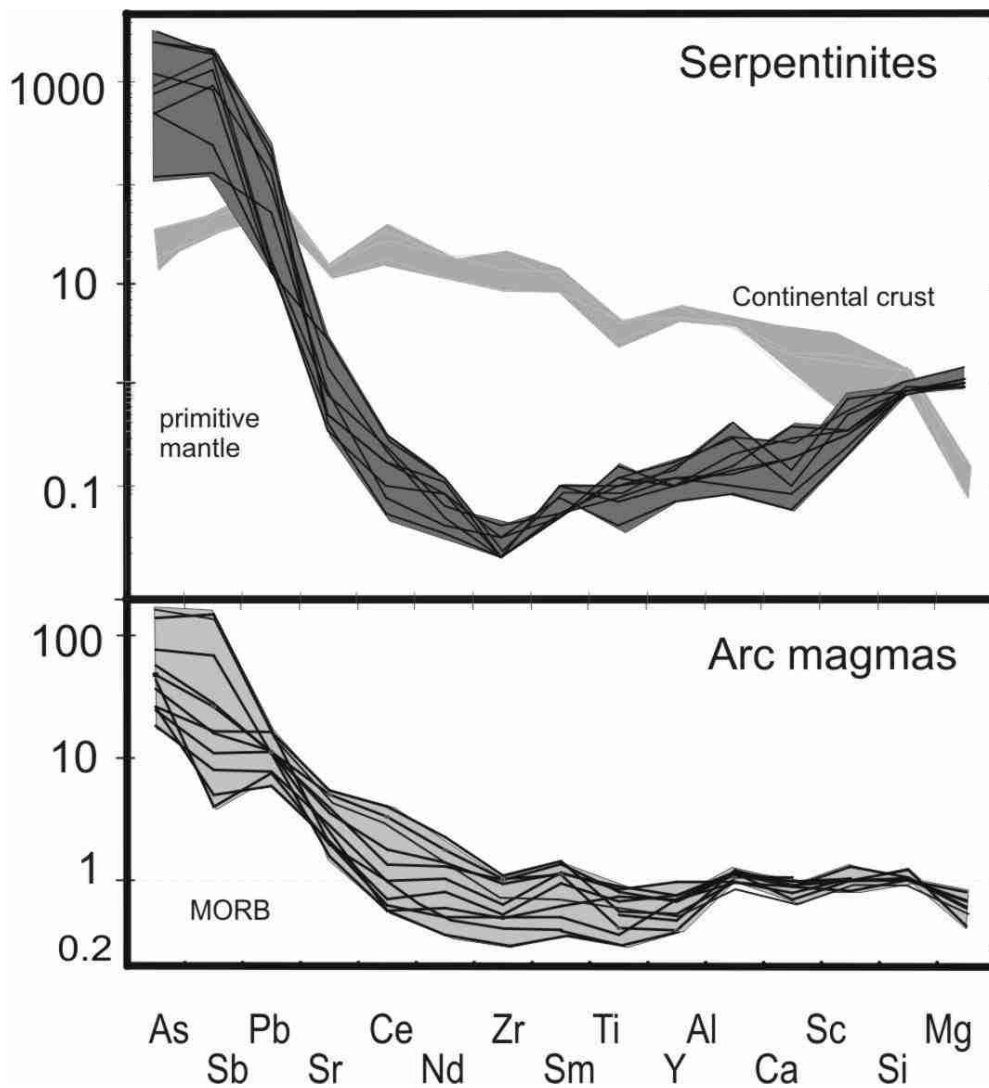


Fig. 1. Composition of serpentinites from the Himalayas compared to those of continental crust and arc magmas. Arsenic and Sb are similar in compatibility to light REE (Noll et al., 1996), but they are placed on the left side of diagram to clarify their fluid-soluble enrichment. Elements from Zr to the right are placed in order of their increasing compatibility with mantle minerals. Serpentinites and continental crust are normalized to primitive mantle (McDonough and Sun, 1995) and arc magmas normalized to mid-oceanic ridge basalts (MORB; Hofmann, 1988; Noll et al., 1996). Arc magmas include data from Pinatubo, Philippines (Bernard, 1997), Esan (Noll et al., 1996), basalt rock standards JB-2 and JB-3 from the Geological Survey of Japan (Ando et al., 1989), volcanic rocks in southern Kyushu (unpublished data by Y. Watanabe and KHH), and Miyake-jima volcano, Japan (deHoog et al., 2002), Telica, Nicaragua (Lefebure, 1986; Noll et al., 1996), and the Medvzhizya and Brat volcanos in the Kuril arc, Russia (Volynets, 1994; Noll et al., 1996). Data for the bulk continental crust and upper and lower crust are from Taylor and McLennan (1995).

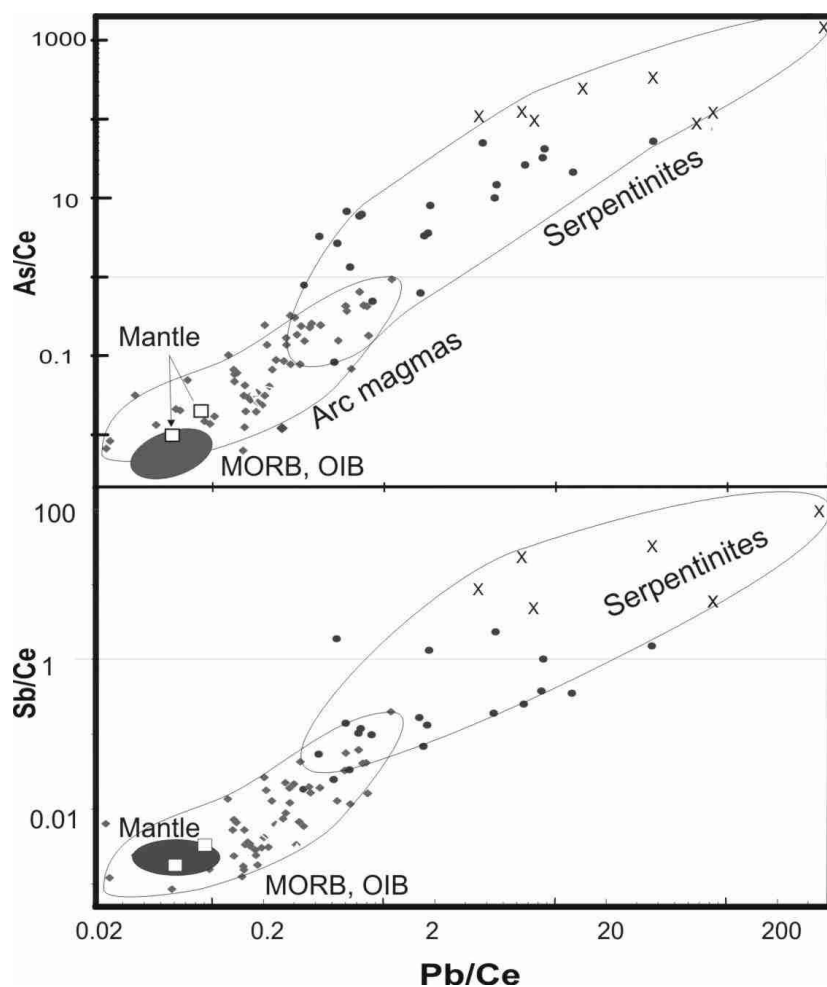


Fig. 2. The concentration ratios of As/Ce, Sb/Ce, and Pb/Ce of serpentinites from the Himalayas (X), Alps (open circles) and Cuba (open triangles). The ratios are compared to those of arc magmas (small solid circles), mantle (open squares for primitive mantle and refractory mantle), oceanic island basalts (OIB) and mid-oceanic ridge basalts (MORB). Ce is mobile in fluids (Fig. 1), but it was selected for the comparison because heavy REE are not commonly determined for many volcanic rocks. The mantle sample with higher As/Ce, Sb/Ce, and Pb/Ce corresponds to primitive mantle (McDonough and Sun, 1995), whereas the sample with low ratios is the depleted mantle (Noll et al., 1996). Data source for arc magmas are listed in Fig. 1, plus mantle (McDonough and Sun, 1995), and those for MORB and OIB (Noll et al., 1996)

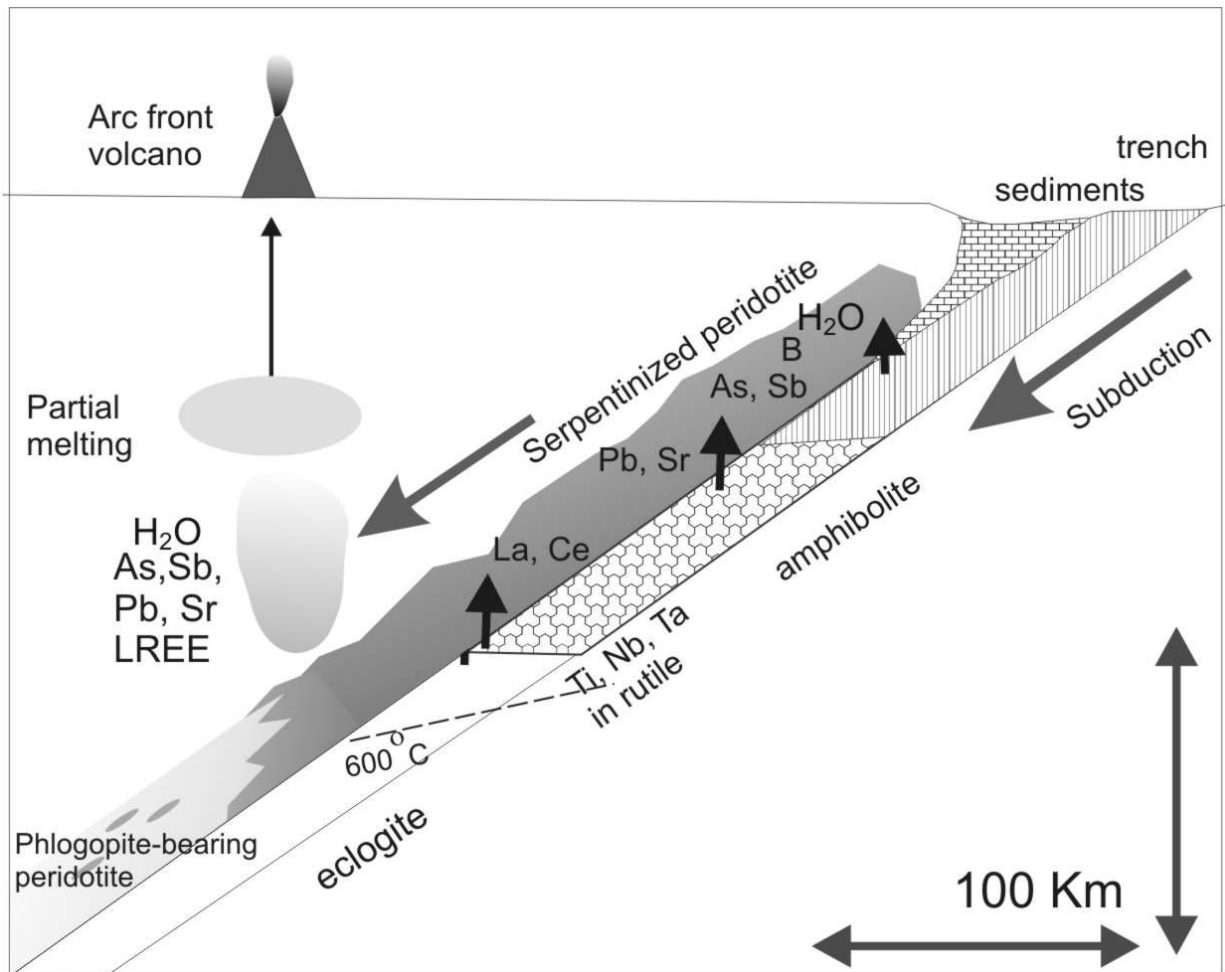


Fig. 3. Mantle peridotites overlying the subduction plane are continuously hydrated during dehydration of slabs, becoming enriched in As, Sb, Pb, Sr and light REE. The serpentinites are transported downward by movement of the subducting slab. When they reach ~ 100 km depth the serpentine minerals break down, releasing most of their H₂O, ~ 13 wt%. The upward migration of the water leads to partial melting that generates arc magmas rich in the fluid-soluble elements enriched in the serpentinite.

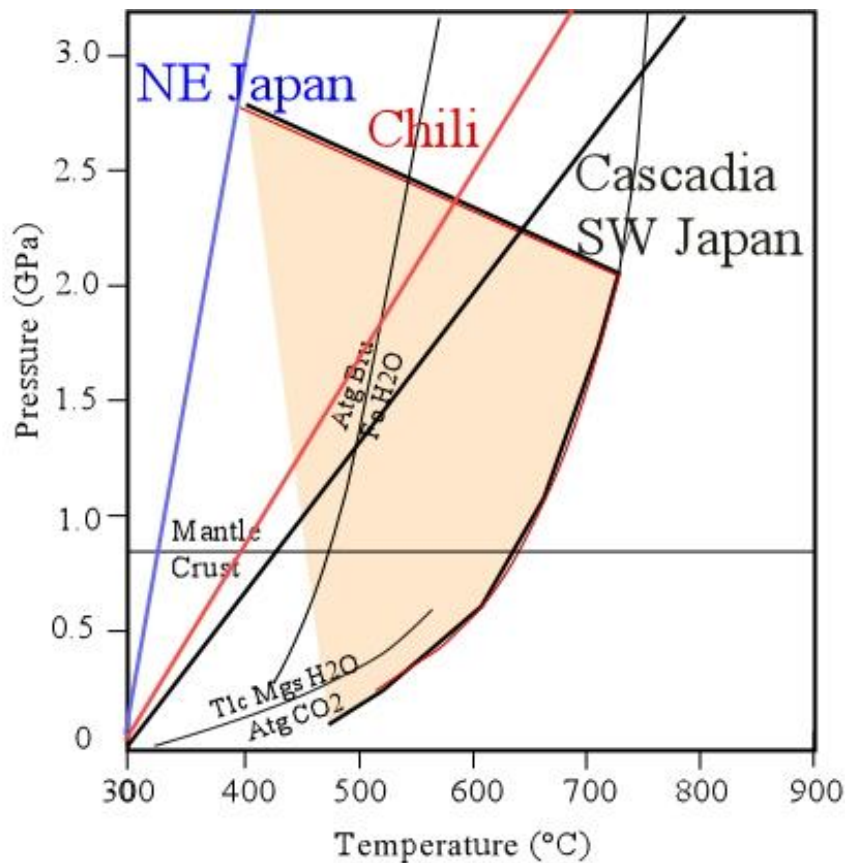


Fig. 3

Fig. 4. Stability of serpentinites along pressure-temperature (P-T) paths of subduction zones (thick solid curves). The PT condition at the top of the 10-km thick serpentinites is schematically shown on the right side of the PT paths for northeastern Japan. The stability of serpentinites is constructed from experimental data (Ulmer and Trommsdorff, 1995; Wunder et al., 2001) combined with invariant points for the dehydration reactions determined by Peacock (2001). Data source for PT paths: Fukaro (1983), Furukawa (1993) and Peacock and Wang (1999).